

**IN THE UNITED STATES PATENT AND TRADEMARK OFFICE**

In re:            Patent Application            : Group Art Unit: 1713  
                    of Matsumoto et al

Serial No.: 10/672,946 : Examiner: M. Bernshteyn

Filed: September 26, 2003 :Attorney Docket  
No.: 8305-233US

For: (Meth)Acrylic (61-0002-1)  
Compound Having An  
Oxetanyl Group And  
Liquid Crystal Film  
Produced By Using  
Same

**DECLARATION UNDER 37 C. F. R. § 1.132**

I, Hitoshi Mazaki, do hereby declare and state as follows:

1. I am the same Hitoshi Mazaki who is a co-inventor and co-applicant in the above-identified patent application. I am at least a co-inventor of all of the presently pending claims.

2. I am a graduate of the Department of Industrial Chemistry, Faculty of Engineering at Tokyo University in March 1985. I also have a degree of Doctor of Engineering for my study of physico-chemical properties of photosynthetic pigments from the same institute, which I received in March 1990.

3. In April 1990, I was employed by Nippon Oil Co., Ltd. Currently, I am employed by Nippon Oil Corporation in the Central Technical Research Laboratory, where I have been actively engaged in the research and development of liquid crystalline polymer films.

4. I have reviewed the final Office Action dated February 13, 2006 in the above-identified application, and copies of U.S. Patent No. 6,136,225 of Meyer et al. ("Meyer") and Hawley's Condensed Chemical Dictionary, 14th Edition, John Wile & Sons, Inc., NY2001 by Richard J. Lewis Sr. ("Lewis"), which the Examiner has used to reject all of the pending claims under 35 U.S.C. §103(a). I am also familiar with the positions that the Examiner has previously taken with regard to the teachings of Meyer and Lewis as they have been applied to the present invention. This Declaration has been prepared to address the issues and concerns raised by the Examiner with respect to Meyer and Hawley.

5. It is my understanding that the Examiner contends that Meyer discloses that  $Z^1$  and  $Z^2$  are independently selected from the groups including acrylic group and epoxy group, and although Meyer does not disclose the specific use of oxetane group, Lewis discloses that an oxetane group ( $=COCH_2C=$ ) is one kind of epoxy group. Furthermore, it is also my understanding that the

6. In order to explain and prove the differences in reactivity between oxetane and epoxy groups, I have carried out additional comparative experiments as set forth below.

I. Synthesis of a benzoic acid derivative having an oxetanyl  
group according to the present invention, "Compound No. 4"

The reaction conditions or the like of each of the steps

are described in detail below.

[Step 1] Synthesis of a methanesulfonyl derivative "Compound No. 2"

Into a 50 ml three-neck flask were charged 7.999 g (68.84 mmol) 3-ethyl-3-hydroxymethyloxetane (OXT-101, manufactured by Toagosei Co., Ltd.), 6.967 g (68.84 mmol) triethylamine, and 30 ml diethylether. A solution of 7.889 g (68.84 mmol) methanesulfonic acid chloride and 15 ml diethylether was added dropwise to the mixture, cooling the flask with ice.

Thereafter, the mixture was cooled with ice for 30 minutes, and then heated to room temperature and reacted for one hour. After the precipitated salt had been filtered out, the diethylether was completely distilled out by vacuum-distillation thereby obtaining 11.783 g of the intended methanesulfonyl derivative, Compound No. 2. The isolated yield was 88.1%.

[Step 2] Synthesis of an ethyl benzoate derivative, "Compound No. 3"

In a 50 ml three-neck flask, 1.662 g (10.00 mmol) ethyl p-hydroxybenzoate, 2.073 g (15.00 mmol) anhydrous potassium carbonate, and 2.349 g (12.09 mmol) the methanesulfonyl derivative obtained in Step 1 were dissolved in 30 ml dimethylformamide.

The solution was heated to a temperature of 100°C as it was turbid and stirred for 4 hours, and then the solid product was filtered out. The dimethylformamide was completely distilled out from the filtrate by vacuum-distillation thereby obtaining the intended oily ethyl benzoate derivative, Compound

No. 3. The resulting compound was used in the next reaction without being further refined.

[Step 3] Synthesis of a benzoic acid derivative, "Compound No. 4"

The ethyl benzoate derivative was placed in an amount of 3.024 g (11.44 mmol, assumed that it was pure) into a flask. Furthermore, 0.77 g (13.72 mmol) potassium hydroxide and 30 ml deionized water were added into the flask. The mixture was heated for 3 hours while being refluxed. After the reaction solution had been cooled, 50 ml ice-cooled deionized water was poured into the flask thereby obtaining a uniform solution.

Thereafter, a solution was separately prepared by dissolving 2.054 g sodium bisulfate monohydrate in 20 ml deionized water. The solution was added slowly dropwise to the solution in the flask.

Since a white crystal precipitated while the sodium bisulfate monohydrate solution was added, it was sufficiently washed with deionized water and then vacuum-filtered. The resulting white crystal was vacuum-dried at room temperature until a constant weight is reached, thereby obtaining 2.092 g the intended benzoic acid derivative having an oxetanyl group, Compound No. 4. The isolated yield was 77.4%.

The  $^{13}\text{C}$  chemical shifts ( $\delta$ ) used for identifying the products obtained in the above syntheses are set forth in Table 1 below.

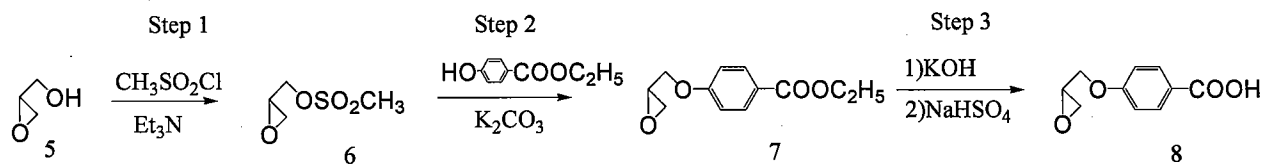
Table 1: Chemical shift of each compound

Assigned Structure	13C NMR chemical shift (ppm)												
	C-1	C-2	C-3	C-4	C-5	C-6	C-7	C-8	C-9	C-10	C-11	C-12	C-13
	8.3	26.4	44.5	78.2		65.2							
	8.1	26.3	43.0	77.4		71.3	37.4						
	8.7	26.9	43.1	77.3		71.1	122.9	115.2	131.9	163.2	166.0	61.0	14.9
	8.6	26.8	43.1	77.3		71.0	123.9	115.0	132.0	163.0	167.7		

## II. Synthesis of a benzoic acid derivative having an epoxy group, "Compound No. 8"

The synthesis of a benzoic acid derivative, "Compound No. 8" was attempted using a compound having an epoxy group as the starting material instead of the compound having an oxetane group used in the above I synthesis.

That is, the procedures of the above I synthesis were followed except that a compound having an epoxy group (glycidol, reagent manufactured by Aldrich) was used as the starting material in place of the compound having an oxetane group (3-ethyl-3-hydroxymethyloxetane (OXT-101, manufactured by Toagosei Co., Ltd.)). The results are set forth below. Scheme B1 below was provided based on the assumption that the same reaction as in I above occurs.



Scheme B1

[Step 1] Synthesis of a methanesulfonyl derivative "Compound No. 6"

Into a 50 ml three-neck flask were charged 5.100 g (68.84 mmol) glycidol (Compound No. 5), 6.967 g (68.84 mmol) triethylamine, and 30 ml diethylether. A solution of 7.889 g (68.84 mmol) methanesulfonic acid chloride and 15 ml diethylether was added dropwise to the mixture, cooling the flask with ice.

Thereafter, the mixture was cooled with ice for 30 minutes, and then heated to room temperature and reacted for one hour. After the precipitated salt had been filtered out, the diethylether was completely distilled out by vacuum-distillation thereby obtaining 4.398 g of the intended methanesulfonyl derivative "Compound No. 6". The isolated yield was 42.0%.

[Step 2] Synthesis of an ethyl benzoate derivative, "Compound No. 7"

In a 50 ml three-neck flask, 1.662 g (10.00 mmol) ethyl p-hydroxybenzoate, 2.073 g (15.00 mmol) anhydrous potassium carbonate, and 1.834 g (12.05 mmol) the methanesulfonyl derivative obtained in Step 1 were dissolved in 30 ml dimethylformamide.

The solution was heated to a temperature of 100°C as it was turbid and stirred for 4 hours and then the solid product was filtered out. The dimethylformamide was completely distilled out from the filtrate by vacuum-distillation thereby obtaining the intended oily ethyl benzoate derivative, Compound No. 7. The resulting compound was used in the next reaction without being further refined.

[Step 3] Synthesis of a benzoic acid derivative, "Compound No. 8"

The ethyl benzoate derivative was placed in an amount of 2.459 g (11.06 mmol, assumed that it is pure) into a flask. Furthermore, 0.77 g (13.72 mmol) potassium hydroxide and 30 ml deionized water were added into the flask. The mixture was heated for 3 hours while being refluxed. After the reaction solution had been cooled, 50 ml ice-cooled deionized water was poured into the flask thereby obtaining a uniform solution.

Thereafter, a solution was separately prepared by dissolving 1.986 g sodium bisulfate monohydrate in 20 ml deionized water. The solution was added slowly dropwise to the solution in the flask.

With the addition of the sodium bisulfate monohydrate solution, brownish powder precipitated and at the same time a small amount of oily phase adhered to the rotors of the magnetic stirrer.

The brownish powder was washed sufficiently with deionized water and recovered by vacuum-filtration and then vacuum-dried at room temperature until a constant weight is reached, thereby obtaining 1.067 g brownish powder. As a result of the analysis of the resulting brownish powder, it was confirmed that the powder was a self-polyaddition product of epoxy and carboxylic acid groups as represented by Compound No. 9 in Scheme B2 shown below.



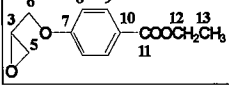
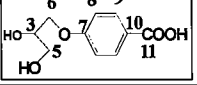
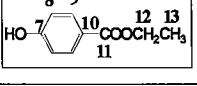
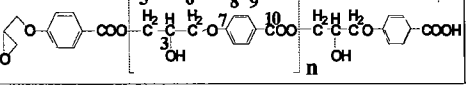
As a result of the  $^{13}\text{C}$ -NMR analysis of the oily phase adhered to the rotors, it was confirmed that the oily phase was predominantly comprised of ethyl hydroxybenzoate as represented by Compound No. 10 in Scheme B2 below.

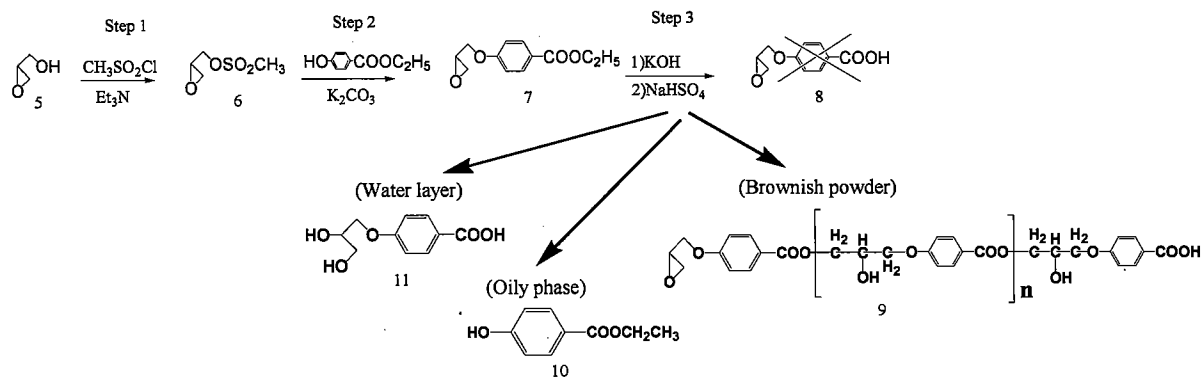


On the other hand, the filtrate (water layer) was fully concentrated by vacuum-distillation thereby obtaining 3.3 g powder (containing an inorganic salt, this inorganic salt does not bother the  $^{13}\text{C}$ -NMR spectrum measurement). It was confirmed that the powder contained a carboxylic acid derivative having a diol group instead of an epoxy group which the intended compound was supposed to have, as represented by Compound No. 11 in Scheme B2 below.

The  $^{13}\text{C}$  chemical shifts of the products obtained in the above syntheses are set forth in Table 2 below.

Table 2: Chemical shift of each compound

Assigned Structure	$^{13}\text{C}$ NMR chemical shift (ppm)												
	C-1	C-2	C-3	C-4	C-5	C-6	C-7	C-8	C-9	C-10	C-11	C-12	C-13
			44.6	-	52.7	62.3							
			44.8	-	49.3	70.5	38.0						
			50.1	-	44.3	69.8	123.0	115.2	131.8	162.6	165.9	60.9	14.8
			70.5	-	63.3	70.4	125.7	114.6	131.8	162.8	168.8		
								115.2	123.0	163.0	166.0	60.9	14.9
			70.5	-	70.0	63.2	124-125	115.0	131.9	162-163	167.8		



Scheme B2

### III. Conclusion

The yield of each step in the above-described experiments are set forth in Table 3 below.

Table 3: Yield in each step

Starting material	Yield in Step 1	Yield in Step 2	Yield in Step 3	Total Yield
Compound 1 (Oxetane compound)	Compound 2 88.1%	Compound 3 99.8%	Compound 4 77.4%	68.1%
Compound 5 (Epoxy compound)	Compound 6 42.0%	Compound 7 97.5%	Compound 8 0%	0%

As apparent from the results set forth in Table 3, the intended benzoic acid derivative, Compound No. 8 was not synthesized when using the starting material, Compound No. 5, having an epoxy group, instead of an oxetane group. It was also confirmed that at least two different kinds of compounds were produced. Therefore, it is now evident from the above results that an epoxy group is completely different in reaction behavior from an oxetanyl group and thus the use of a starting material

having an epoxy group in the above syntheses fails to produce a raw material, i.e., benzoic acid derivative which can be used for synthesizing a (meth)acrylic compound of the present invention.

8. I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Date: June 29, 2006

Hitoshi Mazaki  
Hitoshi Mazaki